Glenn T. Seaborg Center Seminar

Actinide Containing Nanoparticles In The Environment

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Abstract

The migration of low-soluble radionuclides, e.g. Pu(IV), Am(III), Tc(IV), in subsurface conditions is governed by the presence of colloid particles of different nature and composition. However the phenomenon of colloid facilitated radionuclide transport is less studied and is not considered in most of PA calculations.

The presentation summarizes some recent research done at Moscow State University in the field of colloid facilitated transport of actinides. This include the results of (1) UO_2 leaching under 70 ^oC and 150 ^oC under simulated groundwater conditions, (2) XAFS characterization of U(VI) and Th(IV) nanoparticle incorporated in mesoporous silica, (3) nano-SIMS and TEM characterization of colloids collected under different geochemical conditions.

To study the mechanism of actinide nanoparticles formation the laboratory leaching tests of UO_2 under 70 ^oC and 150 ^oC were done in simulated groundwater. The initial sample and solid samples collected from reaction vessels were characterized by SEM, TEM, XPS, EXAFS and XANES. It was established that at 150 ^oC the corrosion of UO_2 is accompanied by formation of colloids of secondary phases that passivate UO_2 surface and prevent its bulk oxidation. In contrast, at 70 ^oC the bulk oxidation of UO_2 is the main mechanism of UO_2 leaching.

In order to study actinide oxide nanoparticle the monodispersed U(VI) and Th(IV) oxide nanoparticles were incorporated in mesoporous structure of MCM-41 and SBA-15 families of SiO_2. The intensity of white line in XANES L_3 spectra and Met-Met interaction was found to be dependant on nanoparticle size.

The radionuclide migration was studied at two nuclear waste sites with oxic (Eh=+100 - +550 mV) and anoxic (Eh= -200- -100 mV) conditions. The colloids were separated by successive micro- and ultra-filtrations and characterized by TEM with EDX and SAED. In order to determine preferential binding of actinides to different colloidal particles the STEM-HAADF and SIMS with submicron resolution was used. Since the concentration of actinides was too small to use XAFS for speciation, the partitioning was studied by sequential extraction.

It was established that in oxic conditions the main part of Pu and Am was bound to nano-colloids while U and Np was mostly found in solution. The main colloidal phases present were: amorphous hydrous ferric oxide >> clays ≈ calcite > rutile ≈ hematite ≈ barite ≈ MnO_2 > monazite >> other phases. According to nano-probe elemental mappings the sequence of actinide binding to different colloidal phases varied in the following manner: amorphous hydrous ferric oxide ≈ MnO_2 > α-Fe_2O_3 while other phases did not sorb actinides.

In anoxic conditions actinides including U and Np are present in tetravalent form and found in colloidal phase. The nano-probe elemental mappings indicate the formation of U(IV) intrinsic colloids that sorb or co-precipitate other actinides.

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